Evidence of Spatially Inhomogeous Pairing on the Insulating Side of a Disorder-Tuned Superconductor-Insulator Transition

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Abstract

Measurements of transport properties of amorphous insulating In_xO_y thin films have been interpreted as evidence of the presence of superconducting islands on the insulating side of a disorder-tuned superconductor-insulator transition. Although the films are not granular, the behavior is similar to that observed in granular films. The results support theoretical models in which the destruction of superconductivity by disorder produces spatially inhomogenous pairing with a spectral gap.

The interplay between localization and superconductivity can be investigated through studies of disordered superconducting films [1], originally treated by Anderson [2], and Abrikosov and Gor'kov [3], who considered the low-disorder regime. Several approaches have been proposed for strong disorder, including fermionic mean field theories [4, 5, 6] and theories that focus on the universal critical properties near the superconductor-insulator transition. The latter consider the transition to belong to the dirty boson universality class [7]. When quantum fluctuations are included in fermionic theories for high levels of disorder a spatially inhomogeneous pairing amplitude is found which retains a nonvanishing spectral gap [8]. For sufficiently disordered systems inhomogeneous pairing can also be brought about by thermal fluctuations [9]. A similar inhomogeneous regime has also been considered under the rubric of electronic microemulsions in the context of the metal-insulator transition of two dimensional electron gases [10]. In this letter we provide evidence of a spatially inhomogeneous order parameter on the insulating side of a superconductor-insulator transition driven by structural and/or chemical disorder.

Studies of disorder and magnetic field tuned superconductor-insulator transitions have usually been carried out on films that are either amorphous or granular. In the former the disorder is on an atomic scale, and in the latter, on a mesoscopic scale in which case the films consist of metallic grains or clusters connected by tunneling, that are either embedded in an insulating matrix, or on a bare substrate [1]. Amorphous films can be produced when films of metal atoms such as Pb or Bi are grown at liquid helium temperatures on substrates precoated with a wetting layer of amorphous Ge or Sb [11], or by careful vapor deposition of Mo_xGe_y, In_xO_y, or TiN using a variety of techniques.

Granular films, are known to develop superconductivity in stages. If the grains are small and weakly connected, the film is an insulator. For grains larger than some characteristic size, and sufficiently close together, "local superconductivity" will develop below some temperature. The opening of a spectral gap in the density of states of the grains [12] results in a relatively sharp upturn in the resistance below this temperature, which is usually close to the transition temperature of the bulk material. For well enough coupled grains, there may be a small drop in resistance at that temperature, followed by this upturn. This is in contrast with the "global superconductivity" that occurs when a sufficient fraction of the grains or clusters are strongly enough Josephson coupled to form a percolating superconducting path across the film.

We have measured the temperature and magnetic field dependence of the resistance, and nonlinear conductance-voltage characteristics of amorphous In_xO_y films prepared by electron-beam evaporation. These films are not granular, but nevertheless exhibit local superconductivity at the lowest temperatures. At low temperatures, the application of a magnetic field results in a dramatic rise in resistance exhibiting a maximum that with decreasing temperatures is found at decreasingly small fields. The conductance-voltage characteristics in this high resistance regime are nonlinear in a manner suggestive of single-particle tunneling between superconductors. We argue that these observations are consistent with the presence of droplets, or islands of superconductivity, characterized by a nonvanishing superconducting pair amplitude and coupled by tunneling. Many of the droplets are Josephson coupled, but their density is not high enough to produce a superconducting path across the film.

The 22 nm thick films used in this study were deposited at a rate of 0.4 nm/s by electron beam evaporation onto (001) $SrTiO_3$ epi-polished single crystal substrates. Platinum electrodes, 10 nm in thickness, were deposited prior to growth. The starting material was 99.999 % pure In_2O_3. A shadow mask defined a Hall bar geometry in which the effective area for four-terminal resistance measurements was 500 x 500 μm^2. As-grown films exhibited sheet resistances of about 4600 Ω at room temperature and about 23 kΩ at 10 K. By annealing at relatively low temperatures (55-70 °C) in a high vacuum environment (10^{-7} Torr), film resistances were lowered, and depending upon the annealing time either insulating or superconducting behavior at low temperatures could be induced

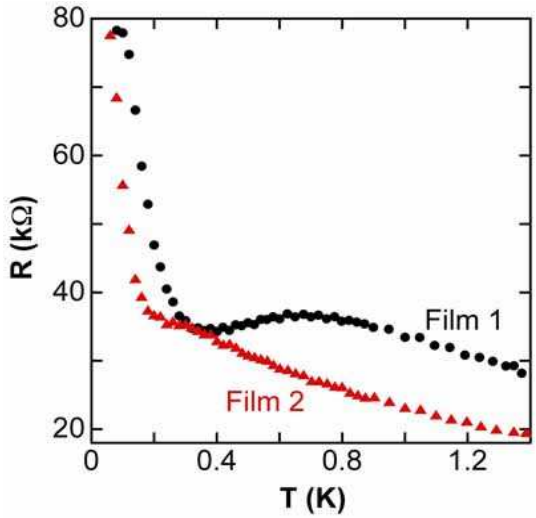

FIG. 1: Resistance vs. temperature for Films 1 and 2.

[13]. Low-temperature rather than high-temperature annealing avoids changes in morphology that would result in granular or microcrystalline films. As reported by Gantmakher, *et al.* [14], at room temperature the resistances of annealed films were found to be unstable. However, at low temperatures (40-1400 mK) and in vacuum, they were stable. The films of the present study had resistances of 2600 Ω at room temperature.

Film structure was studied using atomic force microscopy (AFM), X-ray diffraction (XRD) analysis, and high resolution scanning electron microscopy (SEM). From the XRD there was no indication of the presence of crystalline In or In_2O_3. The SEM did not detect any In inclusions, and could be correlated with AFM studies which revealed for a 22 nm thick film, roughness in the form of surface features with a height of 8.5 nm, and with bases about 18 nm in diameter. The conclusion from these characterization efforts is that the films were homogeneous and amorphous, and did not contain isolated grains or In inclusions.

Measurements were carried out in an Oxford Kelvinox-25 dilution refrigerator housed in a screen room, with measuring leads filtered at room temperature using π-section filters and RC filters. For measurements of resistance, the applied current was set in the range of 10-100 pA, to avoid the possibility of heating. Figure 1 shows a plot of R (T) for two films which were studied in detail. For each, dR/dT is negative at the lowest temperatures. In the case of Film 1 there is a local minimum in $R(T)$ at about 350 mK. Both films exhibit a sharp upturn in $R(T)$ between 200 and 300 mK, with the effects to be discussed below, occurring for Film 1 at higher temperatures than for Film 2. These behaviors are suggestive of a regime of local superconductivity [12].

The sheet resistances of Films 1 and 2 were both approximately 78 kΩ at 40 mK. In a perpendicular magnetic field of only 0.2 T, their sheet resistances increased by up to a factor of 40 at 40 mK. The maximum in $R(B)$ as shown in Fig. 2(a) for Film 1 is followed, at the lowest temperatures, by a relatively slow decrease in resistance with increasing field. The resistance maximum moves to higher fields, with increasing temperature. The behavior of Film 2 resembled the higher temperature data for Film 1, presumably because Film 2 exhibited weaker traces of superconductivity as evidenced by the absence of a local minimum in $R(T)$ in the zero-field. This variation in properties from film to film is expected, as small changes in chemistry and/or morphology can have a large effect on disordered film properties. The temperature dependencies of the fields, B_{peak} and resistances R_{peak} are presented in Fig. 2(b). A qualitatively similar, but weaker enhancement of resistance was previously reported for insulating In_xO_y films by Gantmakher and coworkers [14]. A larger enhancement was reported for ultrathin insulating Be thin films [15]. However neither of these works demonstrated the systematic effects shown in Fig. 2(b).

To probe the nature of the high resistance state, differential conductance-voltage characteristics were also studied [15, 16, 17, 18, 19, 20]. These are shown in Fig. 3 for Film 2 which was studied in detail. Film 1 exhibited qualitatively similar features.

All of the nonlinear conductance-voltage characteristics are reminiscent of the single-particle tunneling characteristics of superconductor-insulator-superconductor (SIS) tunneling junctions. The effects of electron heating are found at voltages well above the observed conductance thresholds [21]. The fact that the low voltage nonlinearities vanish at temperatures above approximately 200 mK, suggests that they are associated with the presence of a nonvanishing pairing amplitude occurring in the disconnected superconducting regions.

We can model these thin disordered films exhibiting spatially inhomogeneous pairing as random networks of tunneling junctions of various (random) levels of conductivity, connecting superconducting clusters imbedded in an insulating matrix. Some of these junctions are superconducting because they are Josephson coupled. As a consequence there are "superclusters," which are aggregates of Josephson coupled smaller clusters that may cover a macroscopic fraction of the film area. Charge may flow through "superclusters" with zero electrical resistance. However, as long as these do not span the film the resistance will be determined by single-particle tunneling. The sheet resistance of the resultant network can be inferred using the following simple argument [22]. Disconnect all of the junctions in the network whose conductance involves single particle tunneling, and then reconnect them one by one in ascending order of resistance. A stage will be reached at which the next junction completes an infinite cluster connecting the ends of the network. Let the normal state resistance of this last junction

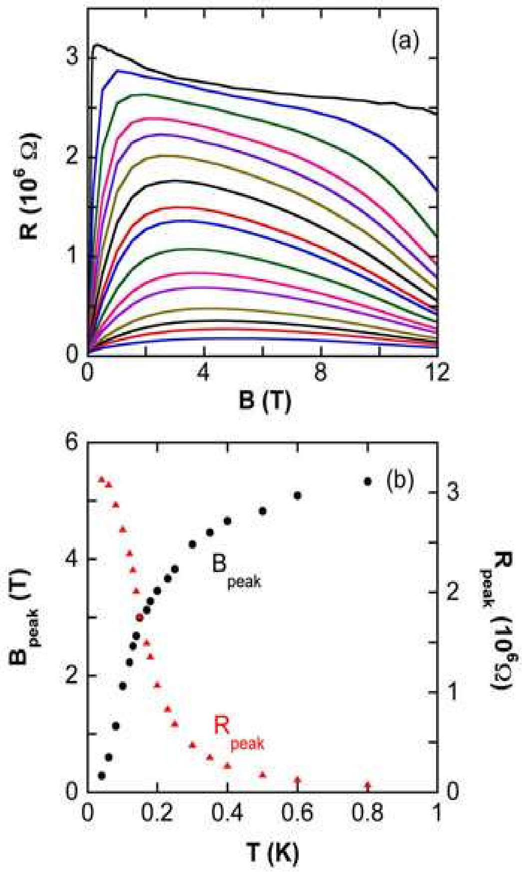

FIG. 2: (a) Resistance vs. magnetic field for Film 1. The temperatures are 40 (top), 80, 100, 120, 130, 140, 150, 170, 180, 200, 230, 250, 300, 350, 400, and 500 mK (bottom). (b)The fields (left axis) and the resistances (right axis) of the peaks in $R(B)$ are plotted as a function of temperature. The flattening of R_{peak} at the lowest temperatures may be the result of a failure to cool the electrons.

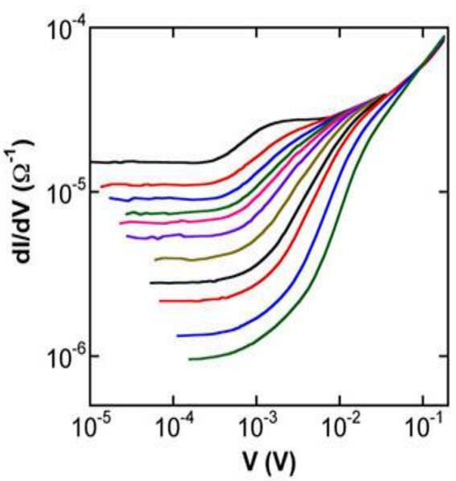

FIG. 3: Differential conductance vs. voltage of Film 2 at 100 mK for 0 (top), 0.01, 0.02, 0.03, 0.04, 0.06, 0.1, 0.175, 0.25, 0.5, and 1 T (bottom).

be R_n. The actual value will depend upon the nature of the distribution of single-particle tunneling resistances in the film. The measured normal-state sheet resistance of the entire network will then be R_n, as this junction is the bottleneck. Junctions with $R > R_n$ are irrelevant since they are always shunted by junctions with resistances of order R_n. Junctions with $R < R_n$ only form finite clusters over macroscopic distances. They don't affect the conductivity because current must still pass through junctions with resistance of order R_n to get from one "supercluster" to the next. The action of a magnetic field is to quench the Josephson coupling within the "superclusters." When this happens, the resistance at each temperature will be governed by the new, higher, value of the bottleneck resistance as there will no longer be any Josephson-coupled "superclusters," and the distribution of junction resistances will shift towards higher values of resistance.

The fact that the magnetic field that induces higher resistance decreases with decreasing temperature is a counter-intuitive result, implying a divergent magnetic length scale in the zero temperature limit, possibly of the form $[\phi_0/B]^{1/2}$where ϕ_0 is the flux quantum. This result suggests enhanced quantum fluctuations in the zero temperature limit. A heuristic argument can be made to demonstrate that this is plausible by treating the inhomogeneous pairing state of the film as a collection of superconducting grains or islands coupled by tunneling junctions. Without the inclusion of quantum fluctuations the argument may not capture all of the features of the data. It is first useful to consider the magnetic field dependence of the in-plane Josephson coupling between two planar thin film square islands with an area L^2. This is a geometry resembling the grain boundary geometry of high temperature superconductor junctions. A magnetic field applied perpendicular to the plane will completely penetrate both electrodes of such a junction. As a consequence the minima of the diffraction pattern will be governed by the field corresponding to a single flux quantum over the full area of the structure or $B\,[L(2L+d)] = \phi_0$ where d is the width of the barrier or gap [23]. Since $L >> d$, the field at the first minimum of the diffraction pattern would be found at a value proportional to ϕ_0/L^{-2} . For a "supercluster" consisting of a square array of square islands that are Josephson coupled, with some degree of randomness in the coupling, one would expect coherence to vanish at the first minimum. For a random array and with clusters that are not square, one might expect a similar dependence on L^{-2}. If the characteristic size of the islands increased with decreasing temperature, which is a plausible assumption, the field quenching the Josephson coupling, would be expected to decrease as is observed. For the films studied, at the lowest temperatures, the peak in the resistance occurs in a field of 0.2 T, which would correspond to a length of approximately 100 nm.

The fall off of the resistance at fields above that producing the maximum slows with decreasing temperature, consistent with the strengthening of the pairing amplitude with decreasing temperature. The fact that this remnant of superconductivity persists to fields up to 12 T, far above the bulk critical field of In_xO_y, implies that the superconducting islands are much smaller than the penetration depth. It should be possible to develop a detailed percolation model of this effect, similar to that developed for granular superconductors [24], which includes the quenching of the Josephson coupling by magnetic field and quantum fluctuations.

Although the resistance of the films of the present work increases with decreasing temperatures in zero magnetic field there is no guarantee that at some temperature lower than the minimum value accessed in these measurements, the resistance will not fall to zero. This could result from the percolation of Josephson coupling across the film as the size of the clusters increases. In that event the inhomogeneous pairing implied by the data would be more likely governed by a theory including both quantum [8] and thermal [9] fluctuations.

The large peaks in the magnetoresistance found at fields above the magnetic field-induced superconductor-insulator transition of superconducting amorphous In_xO_y thin films may result from a similar inhomogeneity of the pairing amplitude, in that case induced by magnetic field rather than disorder. Such peaks were first reported by Hebard and Paalanen [25] who suggested that the state induced when superconductivity was quenched was a Bose insulator, characterized by localized Cooper pairs. They proposed that the peak was the signature of a crossover to a Fermi insulating state of localized electrons. This resistance peak has been the subject of more recent studies of In_xO_y films [14, 16, 26], of microcrystalline TiN films where the high field limit appears to be a "quantum metal" [18], and of high-T_c superconductors [27]. The fact that inhomogeneous pairing can be induced in disordered superconductors by magnetic fields has been recently established using a Hubbard Model [28].

The notion that disorder implies inhomogeneity of superconducting order on some length scale was first discussed by Kowal and Ovadyahu [29], and as was mentioned earlier emerges naturally in a fermionic model of the superconductor-insulator transition that exhibits a disorder-tuned inhomogeneity of the pairing amplitude [8]. The films of Kowal and Ovadyahu differ from those of the present work in that they are presumably more disordered, and thus further into the insulating regime. Their magnetoresistance is always negative as there is no Josephson coupling between islands and the main effect of magnetic field is to weaken the inhomogeneous pairing amplitude, leading to negative magnetoresistance.

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